

Influence of the gate leakage current on the stability of organic single-crystal field-effect transistors

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We investigate the effect of a small leakage current through the gate insulator on the stability of organic single-crystal field-effect transistors (FETs). We find that, irrespective of the specific organic molecule and dielectric used, leakage current flowing through the gate insulator results in an irreversible degradation of the single-crystal FET performance. This degradation occurs even when the leakage current is several orders of magnitude smaller than the source-drain current. The experimental data indicate that a stable operation requires the leakage current to be smaller than 10^{-9} A/cm². Our results also suggest that gate leakage currents may determine the lifetime of thin-film transistors used in applications.

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The study of organic semiconductor transistors aims at the development of organic electronics, for its advantages of being flexible, cheap and suitable for large-area production [1, 2]. So far, considerable research effort has been focused on the optimization of the organic layer to improve the performance of thin-film transistors [3, 4, 5]. Much less attention has been devoted to other important device aspects, such as, for instance, the choice of the gate insulator.

Recent work has demonstrated that the gate insulator plays an important role in determining the device performance [6, 7]. In particular, it has been shown that in polymer as well as in single-crystal organic transistors the mobility of charge carriers is systematically larger the lower the dielectric constant of the gate insulator. This implies that the use of low- ϵ dielectrics will result in a higher device switching speed. In view of this result, it appears useful to investigate systematically how different properties of the gate insulator affect the behavior of organic transistors.

In this paper we use organic single-crystal FETs [8] to investigate how a small leakage current through the gate insulator affects the stability of the device operation. Specifically, we have investigated the behavior of organic single-crystal FETs of different molecules (tetracene, rubrene, perylene) in combination with different dielectrics (Ta₂O₅, ZrO₂, and SiO₂). We find that, irrespective of the specific molecule and dielectric used, leakage current flowing through the gate insulator results in an irreversible degradation of the single-crystal FET operation. The degradation is *not* due to the electrical breakdown of the insulating layer and it also occurs when the leakage current is several orders of magnitude smaller than the source-drain current. From the experimental data, we conclude that a stable operation of organic single-crystal FETs requires the current leaking to the FET channel to be smaller than 10^{-9} A/cm².

The fabrication of the single-crystal FETs used in this work is based on electrostatic bonding of an organic single-crystal to a dielectric surface, with pre-fabricated

source, drain and gate contacts. The details are essentially identical to what has been described in Ref. [9]. Whereas in Ref. [9] only thermally grown SiO₂ was used as gate insulator, here we have also used sputtered layers of Ta₂O₅ and ZrO₂ deposited in different ways. For both Ta₂O₅ and ZrO₂ we have investigated FETs in which the dielectric layers were sputtered from ceramic targets (hereafter referred to as "type I" oxides; see Ref. [10] for details). For Ta₂O₅, we have also investigated the behavior of FETs fabricated on layers sputtered from a metallic target, in the presence of oxygen in the sputtering gas, as described in Ref. [11] (hereafter referred to as "type II" Ta₂O₅). For all FETs discussed here the sputtered oxide layers were approximately 350 nm thick.

The electrical properties of all the different dielectric layers were characterized by capacitance and I - V measurements (see [10]). From these measurements we obtain a dielectric constant $\epsilon = 25$ for Ta₂O₅ (both types) and 23 for ZrO₂, as expected. The breakdown field is comparable for all layers and typically equal to

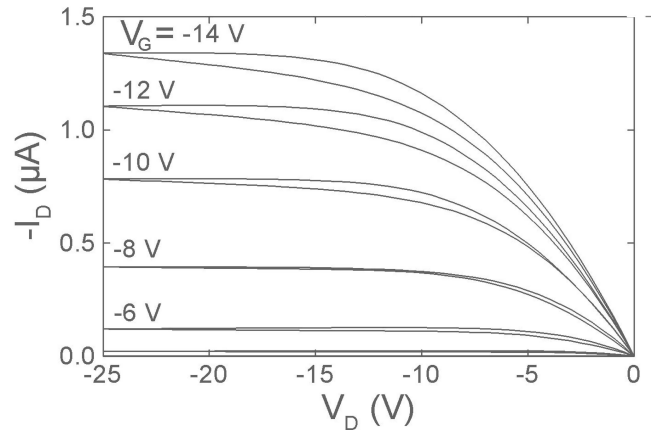


FIG. 1: Current-voltage characteristics of an organic single-crystal FET, with tetracene as the organic molecule and type I Ta₂O₅ as the gate insulator.

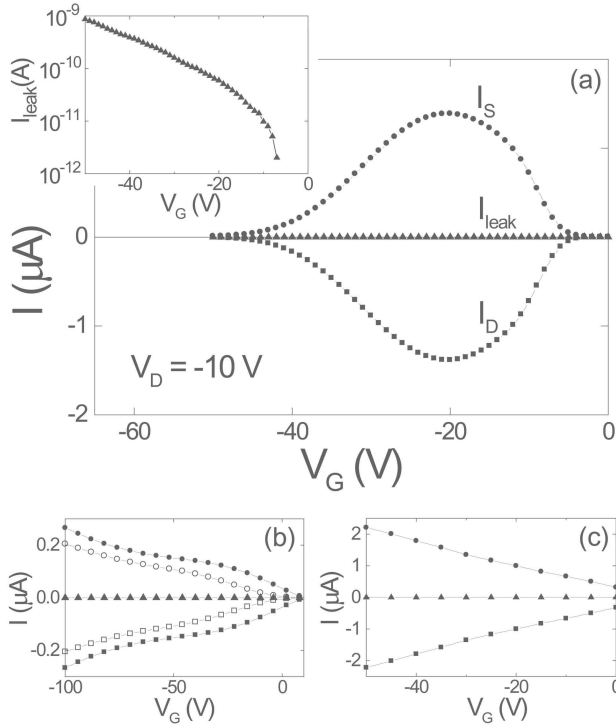


FIG. 2: (a) Transfer characteristics of a FET on Ta_2O_5 ($V_D = -10$ V). The source and drain current (I_S and I_D), are equal and opposite, since the leakage current I_{leak} is orders of magnitude smaller than I_D (see inset). Device degradation is responsible for the non-monotonic I_D - V_G curve, resulting in the full suppression of I_D at high V_G . (b) Transfer characteristics of a tetracene FET on ZrO_2 ($V_D = -10$ V). The I_D - V_G relation is non-linear and I_D and I_S are lower in the second V_G sweep (open circles) than in the first sweep (closed circles). I_{leak} (triangles) is much smaller than I_D and I_S . Note that the shape of the I_D - V_G curves is characteristic for ZrO_2 and differs from that of Ta_2O_5 . For comparison, (c) shows the transfer characteristics of a tetracene FET on SiO_2 for which I_S and I_D are linearly related to V_G ($V_D = -10$ V).

$E_{\text{bd}} \simeq 4 - 6$ MV/cm. The leakage current, on the contrary, is different for the different layers. Specifically, at a voltage corresponding to approximately half the breakdown field, the leakage through type I Ta_2O_5 is typically in the order of 10^{-6} A/cm², slightly higher than that through ZrO_2 , $10^{-7} - 10^{-6}$ A/cm² and much higher than that flowing through type II Ta_2O_5 , $< 10^{-9}$ A/cm² [12].

Electrical characterization of all devices is performed in the dark, at room temperature and under vacuum (10^{-7} mbar), using a HP4156A Semiconductor Parameter Analyzer, in a two-terminal configuration. Fig. 1 shows the transistor characteristics of a tetracene FET with type I Ta_2O_5 as gate dielectric. Upon a superficial inspection, these characteristics resemble those of tetracene FETs on SiO_2 (see Ref. [9]). The typical mobility values in Ta_2O_5 devices range from 0.02 to 0.08 cm²/Vs, smaller than in devices on SiO_2 , as expected in view of the larger dielectric constant of Ta_2O_5 [6]. A first unexpected difference

is however visible in Fig. 1, as the transistor characteristics exhibit some hysteresis, not normally observed in high-quality tetracene single-crystal FETs on SiO_2 . A much more striking difference between the type I Ta_2O_5 and ZrO_2 FETs and the SiO_2 FETs is clearly apparent when looking at the V_G dependence of I_D measured at a fixed V_D value.

Fig. 2a shows the data obtained from a type I Ta_2O_5 FET. Contrary to the usual behavior observed in tetracene FETs on SiO_2 (Fig. 2c), i.e. I_D increasing linearly with V_G , the source-drain current in type I Ta_2O_5 FETs increases, reaches a maximum and then decreases again. The decrease results in the full suppression of the source-drain current, unless dielectric breakdown of the insulator occurs first. This behavior has been observed in all fabricated devices, irrespective of the organic material used (tetracene, rubrene, perylene).

For a tetracene FET on ZrO_2 I_D - V_G curves measured at fixed V_D are shown in Fig. 2b. Again, the I_D - V_G relation is markedly non-linear, although full suppression of the source-drain current is not reached. The shape of the non-linearity is different from that observed in type I Ta_2O_5 devices and is characteristic for our ZrO_2 transistors. Also for ZrO_2 FETs, the behavior of the I_D - V_G is similar when crystals of different organic materials are used.

The anomalous behavior reproducibly exhibited by type I Ta_2O_5 and ZrO_2 FETs originates from *irreversible device degradation*. Specifically, we observe that, for every device studied, repeating the measurement of the I_D - V_G curve systematically results in lower measured values of I_D (see Fig. 2b). For those type I Ta_2O_5 transistors in which the increase in V_G is sufficient to fully suppress the source-drain current (see Fig. 2a), no field-effect induced current is ever observed after the measurement, indicating that the degradation of the device is complete.

Inspection of the degraded transistors using an optical microscope does not reveal any visible change in the device. The bulk of the crystal, the dielectric layer, and the FET circuitry appear to have all remained intact and the crystal is still well bonded to the substrate. This suggests that the device degradation is confined to the first layers of the organic material at the interface with the dielectric.

To determine the cause of device degradation it is revealing to compare the behavior of single-crystal FETs fabricated using type I and type II Ta_2O_5 . Fig. 3 shows the results of I_D - V_G sweeps for two rubrene single-crystal FETs fabricated using the two different oxides. Similar to what we have shown for the tetracene FETs in Fig. 2, the FET on type I oxide exhibits a non-linear, non-monotonic I_D - V_G relation up to the dielectric breakdown voltage (at $V_G \approx -80$ V for this sample). Conversely, for the rubrene FET on type II Ta_2O_5 , I_D scales linearly with V_G in a large range of values above the threshold voltage, up to the maximum voltage reached in the experiment ($V_G = -100$ V, corresponding to a charge density of $\simeq 5 \times 10^{13}$ holes/cm²). For this FET, multiple mea-

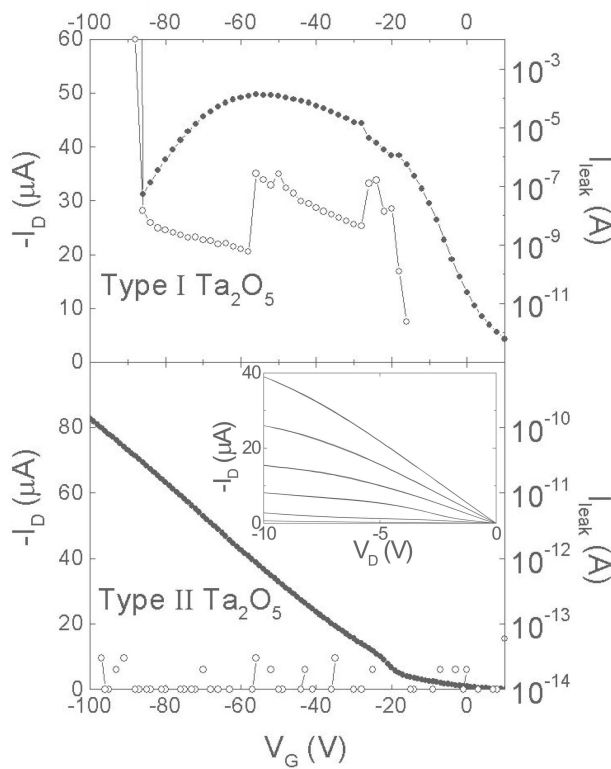


FIG. 3: I_D - V_G curves (closed symbols) of two rubrene FETs on type I Ta_2O_5 (upper panel) and type II Ta_2O_5 (lower panel) ($V_D = -10$ V in both cases). The open symbols represent the leakage current. The degradation-induced, non-monotonic behavior of the I_D - V_G curve observed for the type I Ta_2O_5 FET is absent in the type II Ta_2O_5 device. Note that the leakage current in the two devices differs by orders of magnitude, despite the comparable crystal surface area ($\sim \text{mm}^2$) and Ta_2O_5 thickness (~ 350 nm). The inset shows that FETs on type II Ta_2O_5 exhibit hysteresis-free electrical characteristics.

measurements of the I_D - V_G curve reproducibly give the same result. Note also, in the inset of Fig. 3, that the transfer characteristics of the rubrene FET on type II Ta_2O_5 are fully hysteresis-free, as is also typical for high-quality transistors fabricated on SiO_2 . In short, contrary to what happens to devices based on type I Ta_2O_5 , for FETs fabricated on type II Ta_2O_5 device degradation does not occur. Since the main difference between type I and type II Ta_2O_5 layers is the much higher level of leakage current observed in the type I layers, this observation suggests that the current leaking through the gate insulator is the cause for the device degradation.

To further investigate the origin of the FET degra-

dation, we have also studied FETs fabricated on bilayers consisting of a 350 nm thick layer of type I Ta_2O_5 (ZrO_2) covered with a 25 nm thin top layer of ZrO_2 (type I Ta_2O_5), so that the organic crystal is in contact with the thin top layer. For these FETs, the shape of the I_D - V_G curve is similar to that observed in FETs where the thin top layer is not present. These experiments indicate that the details of the device degradation are determined by the thick oxide layer and not by the material directly in contact with the organic crystals. This observation rules out the possibility that a chemical reaction between molecules and dielectric material is causing the device degradation and confirms the role of the leakage current, since in these oxide bi-layers it is the thick layer that determines the magnitude of I_{leak} .

We conclude that damage to the organic crystal induced by current leaking through the gate insulator is the cause for the device degradation [13]. This conclusion is further supported by the absence of degradation in single-crystal FETs fabricated on SiO_2 , in which the leakage current is undetectably small. It is also consistent with the larger degradation observed in type I Ta_2O_5 FETs as compared to ZrO_2 devices, since the leakage current through ZrO_2 is typically almost an order of magnitude less than in type I Ta_2O_5 .

It is worth noting that degradation occurs even when the leakage current is several orders of magnitude lower than the source-drain current. Specifically, our data quantitatively show that in organic single-crystal FETs gate leakage currents larger than approximately 10^{-9} A/cm² [14] systematically result in irreversible device degradation. This conclusion poses a clear constraint on the design of properly functioning single-crystal FETs. It is possibly also relevant for organic thin-film transistors, as it suggests that the gate leakage current is an important factor in determining the device lifetime.

In conclusion, we have shown that leakage current from the gate electrode causes irreversible degradation of organic single-crystal FETs, even when it is orders of magnitude smaller than the source-drain current. This poses a clear constraint for the design of single-crystal transistors currently used to investigate the intrinsic electronic properties of organic semiconductors. As a by-product of this work, we have successfully fabricated single-crystal devices operating up to a charge density of at least 5×10^{13} carriers/cm² (~ 1 carrier per 10 molecules), which will enable the investigation of organic single-crystal FETs at high carrier density.

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- [12] The microscopic origin of the difference in leakage current between Type I and Type II Ta₂O₅ is probably due to the larger deposition rate that can be achieved in sputtering from a metal target, which results in the inclusion of less impurities in the sputtered layers.
- [13] The specific microscopic process responsible for the degradation of the organic material remains to be understood. One possible mechanism is that high-energy electrons leaking through the gate insulator physically break individual molecules at the crystal surface, thus causing the appearance of a very large number of traps in the FET active regions.
- [14] A more precise quantification is difficult because an unknown fraction of leakage current flows directly to the source or drain contacts and does not cause damage to the organic crystal.